A new constitutive model for carbon-black reinforced rubber in medium dynamic strains and medium strain rates

F. Carleo
Division of Engineering Science, Soft Matter Group, School of Engineering and Materials Science Materials Research Institute, Queen Mary University of London, Mile End Rd, London, E1 4NS, UK

J.J.C. Busfield
Soft Matter Group, School of Engineering and Materials Science, Materials Research Institute Queen Mary University of London, Mile End Rd, London, E1 4NS, UK

R. Whear
Jaguar Land Rover Ltd, Banbury Road, Gaydon, Warwick, CV35 ORR

E. Barbieri
Division of Engineering Science, School of Engineering and Materials Science, Materials Research Institute Queen Mary University of London, Mile End Rd, London, E1 4NS, UK

ABSTRACT: Modelling the viscoelastic behaviour of rubber for use in component design remains a challenge. Previous reviews (Diani, Fayolle, & Gilormini 2009) and our studies presented in this paper highlight the issues of using of the most common viscoelastic non-linear constitutive models (Besdo & Ihlemann 2003; Bergström & Boyce 1998;Ogden & Roxburgh 1999). In detail, such models cannot reproduce or predict the experimental stress data for filled natural rubber loaded under the typical operating conditions. Examples of such conditions include cyclic strain history with constant strain rates and variable amplitude. This paper examines the behaviour of natural rubber elastomers filled with different percentages of carbon black. The elastomers chosen are typical of the materials used in vibration damping or automotive suspensions. We show that a constitutive model based on the fractional calculus can provide a good agreement for cyclic uniaxial tensile tests at a constant amplitude. The proposed model can capture, for example, the hysteresis and cyclic stress softening observed in the experimental data.

1 INTRODUCTION

The research on the viscoelastic properties of rubber-like materials has attracted much attention particularly in the automotive sector. Two different philosophies exist in formulating constitutive models: the phenomenological and the mechanistic approach. The first is based on direct observation, on the measure and curve-fitting of the experimental data, the second consists in a derivation of the strain energy function from statistical considerations on the molecular motion of polymeric chains. Notwithstanding a significant number of the non-linear viscoelastic models proposed in the literature, previous reviews (Diani, Fayolle, & Gilormini 2009) and our studies show that none of them can fit the real behaviour of filled rubber.

The existing models are each developed under specific loading conditions. For quasi-static applications, numerous and well-established hyperelastic models exist in the literature. An hyperelastic constitutive law consists of an equation relating the strain energy density to the three invariants of the strain tensor. These models can predict with few material parameters the rubber behaviour in tension, shear, compression or biaxial load. Also, these models are the basis for numerous models that try to predict the viscoelastic response. For dynamic applications, the distinction between linear and non-linear viscoelastic theories is important. Filled rubber has a highly non-linear viscoelastic behaviour. Different models for different loading condition exist that account for the high non-linearity of the filled vulcanised rubber, but no current constitutive law can model all the non-linear viscoelastic effects that the filled rubbers exhibit in the regime of interest.

The phenomenological models can roughly be classified into two large groups: the damage models and the rheological models with serial and parallel combination of elastic and viscous elements.

The damage models (Ogden & Roxburgh 1999, Dorfmann & Ogden 2003) are not able to model the different loading and reloading stress-strain path with the consequence that they fail to predict the cyclic stress softening phenomenon.

Rheological models (Bergström & Boyce 1998, Hurtado, Lapczyk, & Govindarajan 2013) with elastic and viscous component require too many calibration parameters that change with the experimental input, hence these models are not predictive.

This paper presents a new constitutive model able to represent the mechanical response of a filled rubber loaded with cyclic strain history at medium strain amplitude. Section 2 presents the experimental campaign. Cyclic uniaxial tensile tests were carried to take a close look at the nonlinear behavior of the vulcanized Natural Rubber (NR) filled with different amount of carbon black. Section 3 shows the fractional calculus method and its use in formulating the proposed constitutive model. In particular, section 3.1 reviews fractional calculus in the context of nonlinear viscoelasticity. The new model is presented in section 3.2. Sections 4 and 5 present the preliminary results, the limitations of the actual model and future works.

2 EXPERIMENTAL TESTS

Cyclic uniaxial tensile tests were carried out to examine the nonlinear behavior of the vulcanized Natural Rubber (NR) filled with different amounts of carbon black. In addition, the two separate strain history inputs adopted (Figure 1 and Figure 2) allow analysis of phenomenon such as hysteresis, cyclic stress softening, recovery, permanent set and pre-strain effect.

In the range where automotive components are expected to operate, the filled vulcanised elastomers display high nonlinearity, large hysteresis, Mullins effect and permanent set.

The hysteresis increases with the amount of carbon black in the compound.

There is a reduction in the stress on each successive loading at the same strain amplitude. The decline is largest in the first and second loading-unloading cycles and becomes rather small in the following cycles (Figure 4). The effects of stress softening are not a major issue for a compound with low content of CB.

The behaviour of rubber is rate dependent, and there is an enhancement of stress when the deformation rate is increased.

The unloading and reloading responses differ.

The material tries to return to the virgin loading path whenever the load increases beyond its previous maximum value, after a transient behaviour. The

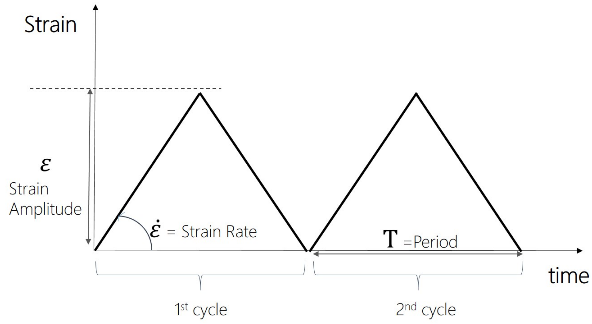

Figure 1: Schematic layout: Cyclic strain history with constant strain amplitude and strain rate

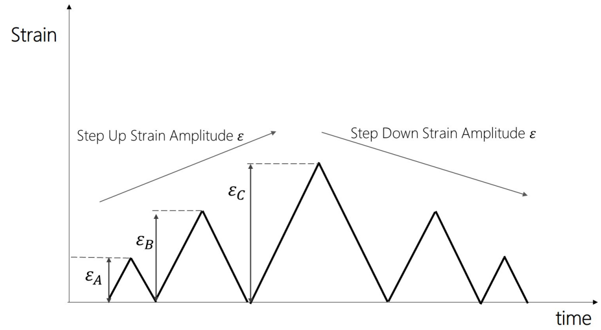

Figure 2: Schematic layout: Cyclic strain history with constant strain rate and various strain amplitude organised in step up and step down.

stress-strain path is highly dependent on the loading history (Figure 3).

3 FRACTIONAL CALCULUS (FC) METHOD

3.1 *Literature review*

Fractional Calculus (FC) is a branch of mathematics that generalizes the operators of differentiation and integration from integer to fractional order. Some important properties are:

- the operator is linear;
- the zero order derivative of a function returns to the function itself;

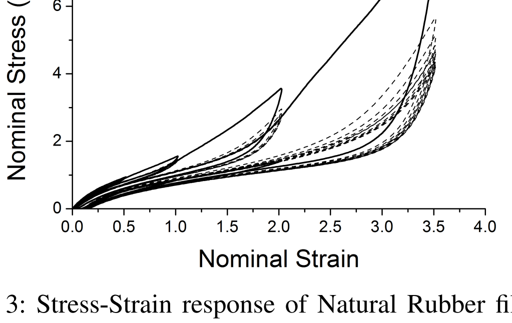

Figure 3: Stress-Strain response of Natural Rubber filled with 30phr of carbon black (NR30) submitted to cyclic uniaxial tension with a strain history input as in figure 2.

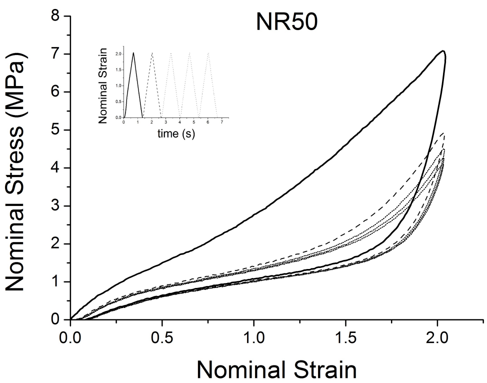

Figure 4: Stress-Strain response of Natural Rubber filled with 50phr of carbon black (NR50) submitted to cyclic uniaxial tension with a strain history input as in figure 1.

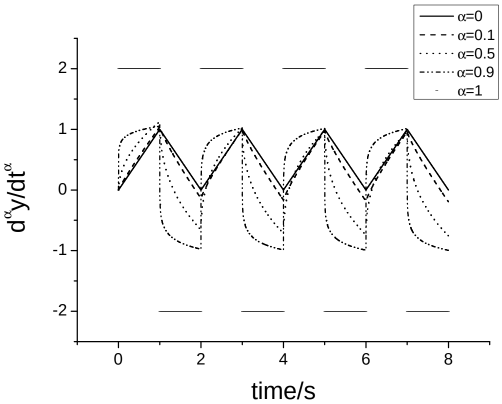

Figure 5: Fractional derivative of a triangular function

- when the order is an integer, FC gives the same results of the ordinary operation.

In the literature, different definitions are available for the fractional derivative (FD), such as the Riemann-Liouville, Caputo or Grunwald- Letnikow (Eq.1).

$$D_x^\alpha f(x) = \lim_{h \longrightarrow 0} \frac{1}{h^\alpha} \sum_{j=0}^{\frac{x}{h}} (-1)^j \binom{\alpha}{j} f(x - jh) \qquad (1)$$

These operators are not a merely mathematical curiosities but have numerous applications in different fields. One example is the study of materials containing elastic and viscous components.

The FD method is an interpolation between elastic and viscous behaviour. The elastic material (spring) connects the stress with the strain (zero order derivative), while in the viscous material the stress is proportional to the first time derivative of the strain. In the rheological models, this component is called a springpot.

A justification for the use of the empirically developed FC models exists. Bagley & Torvik (1983) proved a link between molecular theories and FC approach, showing that Rouses molecular theories describing viscoelasticity produced a fractional derivative relationship between stress and strain.

In the literature, the applications of fractional derivative models are numerous in linear viscoelastic problems such as creep and relaxation (Schmidt & Gaul 2001; Pritz 2003; Sasso, Palmieri, & Amodio 2011; Zhou, Wang, Han, & Duan 2011). Linear fractional derivative models, such as Fractional Kelvin or Fractional Maxwell, are popular because they describe the behaviour of viscoelastic dampers with a small number of parameters.

Several attempts to model the nonlinearity have also been made (Sjöberg & Kari 2003; Ramrakhyani, Lesieutre, & Smith 2004), particularly in the case of cyclic loading condition with small strain amplitude, where the filled rubber shows a dependence of the viscoelastic storage modulus on the magnitude of the applied strain. This effect is known as the Fletcher-Gent effect or the Payne effect (Lion & Kardelky 2004).

The main advantage is that fractional models describe the behaviour of viscoelastic materials with a small number of parameters. The drawback is that it is difficult to estimate the parameters of the model, which are highly dependent on the choice of the first attempt solutions (Lewandowski & Chorażyczewski 2010).

3.2 *A model for non linear viscoelasticity*

The idea to use the fractional derivative to describe phenomena at medium strain amplitude derives from the ability of this operator to reproduce hysteresis loops for a cyclic strain history. Figure 5 shows various orders of fractional derivatives of a triangular function.

The total stress in the present paper is obtained by adding to a viscous damaging part an hyperelastic stress (Eq. 2).

The formulation of this new model is a modification of the Nonlinear Fractional Voigt model. The basic behaviour is well represented by an hyperelastic model as the Yeoh theory (Eq. 3, for uniaxial tension test). The remaining part of the total stress is obtained multiplying the fractional derivative of the strain with a linear function expressed in equation 4 as the strain input function.

$$\sigma = \sigma_H + \sigma_{FC} \qquad (2)$$

$$\sigma_H = 2\left(C_1 + 2C_2\left(I_1 - 3\right) + 3C_3\left(I_1 - 3\right)^2\right) \qquad (3)$$

$$\left(\lambda^2 - \frac{1}{\lambda}\right)$$

$$\sigma_{FC} = \varepsilon(t) \cdot D_t^\alpha \varepsilon(t) \qquad (4)$$

This formulation has three parameters for the hyperelastic part (C_1,C_2,C_3) and one parameter (α) for the order of the fractional derivation. The model uses two different orders of derivation. One order of derivation needs to define the primary loading path. A second value of α is used to fit the first unloading curve and the following cycles. The model in total needs just 5 parameters.

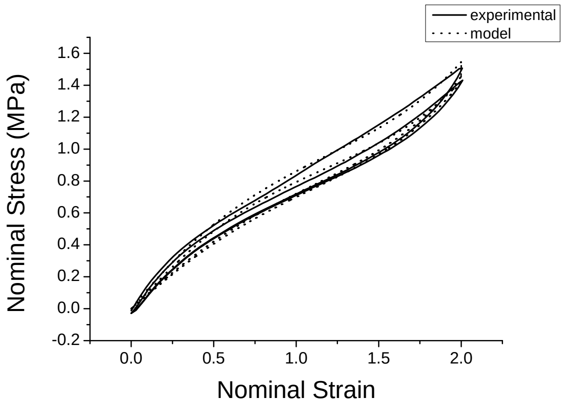

Figure 6: Best-fit curves: comparison of calculated and experimental data for NR10

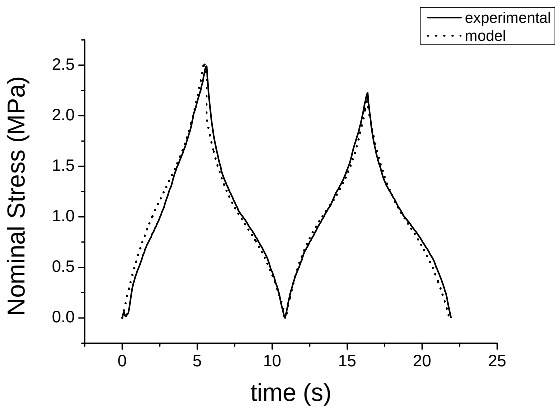

Figure 7: Best-fit curves: comparison of calculated and experimental data for NR20

4 PRELIMINARY RESULTS AND LIMITATIONS

The parameters are determined during an optimisation algorithm that minimises the mean square error. Figures 6, 7 and 8 show the comparison of experimental and calculated data.

For compounds with a small amount of carbon black, the model reproduces qualitatively the first loading and the relaxed cycle. However, the theoretical first unloading curve is unable to reproduce the high tangent modulus that the experimental data show at high strain amplitude.

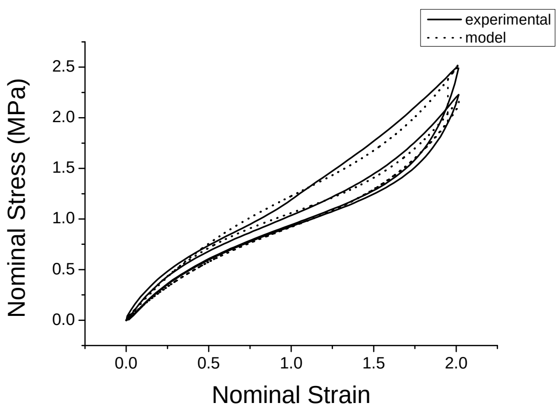

Figure 8: Best-fit curves: comparison of calculated and experimental data for NR20

5 CONCLUSIONS AND FUTURE WORKS

The model described with use of FC provides a good fitting, requires only few parameters, and the approach is physically justified (Bagley & Torvik 1983). Future improvements on the model are expected to define the best fitting in the first unloading path at high strain amplitude. Also, the model has to be able to predict the behaviour of other compounds with a higher amount of carbon black and a higher nonlinear viscoelastic behaviour. Furthermore, the model will be tested on more complex strain history.

REFERENCES

Bagley, R. L. & P. Torvik (1983). A theoretical basis for the application of fractional calculus to viscoelasticity. *Journal of Rheology 27*(3), 201–210.

Bergström, J. & M. Boyce (1998). Constitutive modeling of the large strain time-dependent behavior of elastomers. *Journal of the Mechanics and Physics of Solids 46*(5), 931–954.

Besdo, D. & J. Ihlemann (2003). A phenomenological constitutive model for rubberlike materials and its numerical applications. *International Journal of plasticity 19*(7), 1019–1036.

Diani, J., B. Fayolle, & P. Gilormini (2009). A review on the mullins effect. *European Polymer Journal 45*(3), 601–612.

Dorfmann, A. & R. Ogden (2003). A pseudo-elastic model for loading, partial unloading and reloading of particle-reinforced rubber. *International Journal of Solids and Structures 40*(11), 2699–2714.

Hurtado, J., I. Lapczyk, & S. Govindarajan (2013). Parallel rheological framework to model non-linear viscoelasticity, permanent set, and mullins effect in elastomers. *Constitutive Models for Rubber VIII 95*.

Lewandowski, R. & B. Chorażyczewski (2010). Identification of the parameters of the kelvin–voigt and the maxwell fractional models, used to modeling of viscoelastic dampers. *Computers & structures 88*(1), 1–17.

Lion, A. & C. Kardelky (2004). The payne effect in finite viscoelasticity: constitutive modelling based on fractional derivatives and intrinsic time scales. *International Journal of Plasticity 20*(7), 1313–1345.

Ogden, R. & D. Roxburgh (1999). A pseudo–elastic model for the mullins effect in filled rubber. In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, Volume 455, pp. 2861–2877. The Royal Society.

Pritz, T. (2003). Five-parameter fractional derivative model for polymeric damping materials. *Journal of Sound and Vibration 265*(5), 935–952.

Ramrakhyani, D. S., G. A. Lesieutre, & E. C. Smith (2004). Modeling of elastomeric materials using nonlinear fractional derivative and continuously yielding friction elements. *International journal of solids and structures 41*(14), 3929–3948.

Sasso, M., G. Palmieri, & D. Amodio (2011). Application of fractional derivative models in linear viscoelastic problems. *Mechanics of Time-Dependent Materials 15*(4), 367–387.

Schmidt, A. & L. Gaul (2001). Fe implementation of viscoelastic constitutive stress-strain relations involving fractional time derivatives. *Constitutive models for rubber 2*, 79–92.

Sjöberg, M. & L. Kari (2003). Nonlinear isolator dynamics at finite deformations: an effective hyperelastic, fractional derivative, generalized friction model. *Nonlinear Dynamics 33*(3), 323–336.

Zhou, H., C. Wang, B. Han, & Z. Duan (2011). A creep constitutive model for salt rock based on fractional derivatives. *International Journal of Rock Mechanics and Mining Sciences 48*(1), 116–121.